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Physical-Chemical Nitrogen Removal Wastewater Treatment

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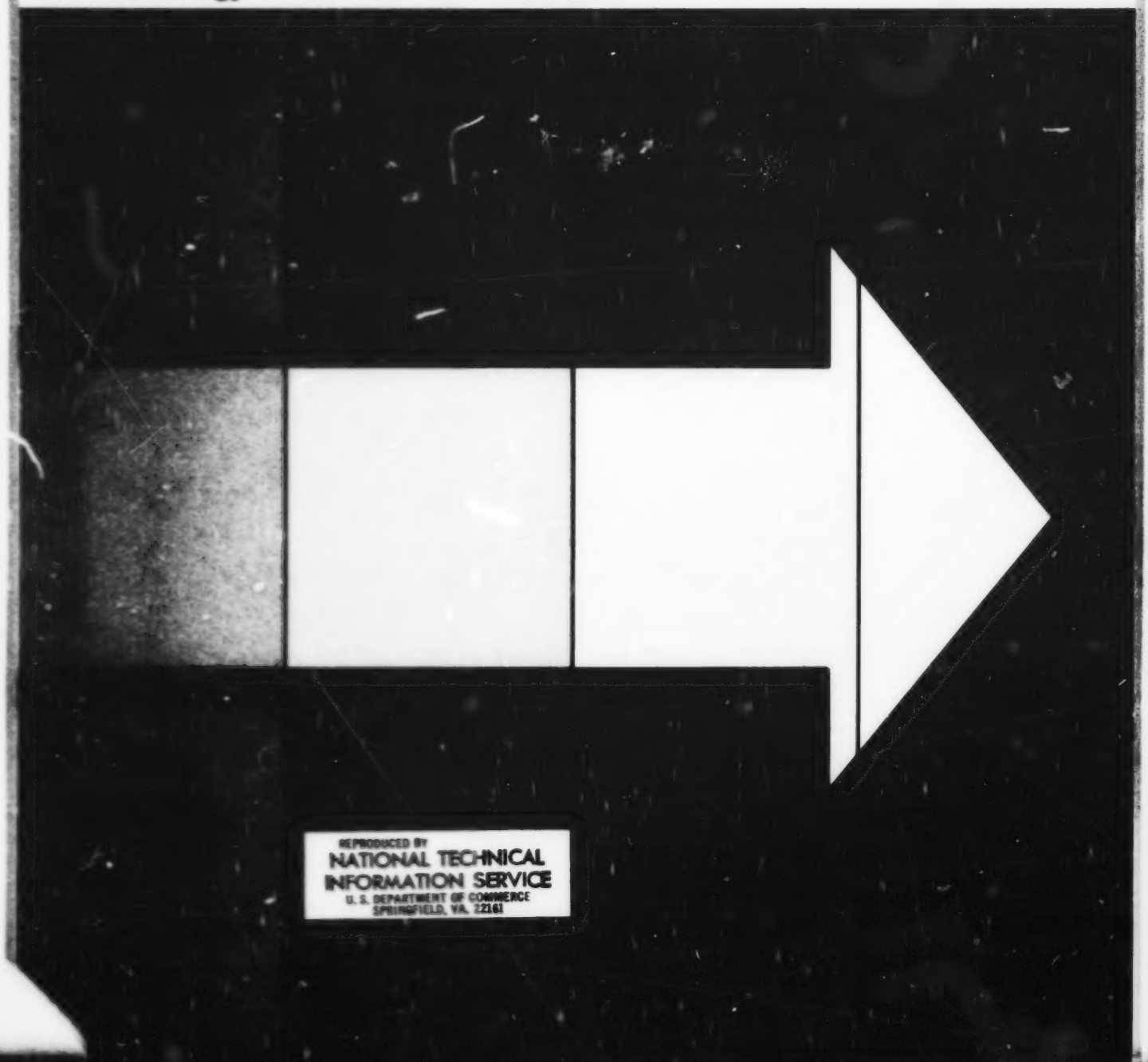

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July 1974

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INTRODUCTION

There are three basic physical-chemical nitrogen-removal techniques available for application today. These three processes are

- Ammonia stripping (ch. I)
- Selective ion exchange (ch. II)
- Breakpoint chlorination (ch. III)

All of these approaches have the advantage that they are based on the removal of nitrogen in ammonia form, which eliminates the costs of converting the ammonia to nitrate in the biologic-treatment step. They also have the advantages that they are unaffected by toxic compounds that can disrupt the performance of a biologic nitrogen-removal system, they are predictable in performance, and the space requirements for the treatment units are less than for biologic-treatment units.

The advantages and disadvantages of each of these physical-chemical processes are discussed in detail and the processes are compared in the chapters that follow. Discussion of these processes includes application at the following facilities, either in existence or under design:

- South Lake Tahoe, Calif.
- Orange County, Calif.
- Windhoek, South Africa
- Blue Plains, D.C.
- Upper Occoquan Sewage Authority, Va.
- Rosemount, Minn.
- North Lake Tahoe, Calif.
- Montgomery County, Md.
- Cortland, N.Y.

Chapter I

AMMONIA STRIPPING

The only nitrogen-removal process that actually has been used on a plant scale in wastewater treatment is ammonia stripping. This process has been in use for ammonia nitrogen at the South Lake Tahoe plant for about 4 years. Both the advantages and limitations of this process have been clearly demonstrated.

The ammonia-stripping process itself consists of

- Raising the pH of the water to values in the range of 10.8 to 11.5, generally with the lime used for phosphorus removal
- Formation and re-formation of water droplets in a stripping tower
- Providing air-water contact and droplet agitation by circulation of large quantities of air through the tower

The towers used for ammonia stripping closely resemble conventional cooling towers.

Questions are sometimes raised concerning the fate of ammonia discharged to the atmosphere. Are we merely converting a water-pollution problem to an air-pollution problem? Does the ammonia stripped from the wastewater cause an air-pollution problem or find its way back to the receiving stream owing to scavenging by precipitation?

The concentration of ammonia in the stripping-tower discharge is only about 6 mg/m^3 for domestic wastewaters (at an air flow of 500 ft^3/gal and at an ammonia concentration of 23 mg/l in the tower influent). As the odor threshold of ammonia is 35 mg/m^3, the process does not present a pollution problem in this respect. The ammonia discharged to the atmosphere is a stable material that is not oxidized to nitrogen oxides in the atmosphere. The natural production and release of ammonia as part of the natural nitrogen cycle is about 50 billion tons per year. Roughly 99.9 percent of the atmosphere's ammonia concentration is produced by natural biological processes.[1] There is a large turnover of ammonia in the atmosphere, with the total ammonia content being displaced once a week on the average. Ammonia is returned to the earth through gaseous deposition (60 percent), aerosol deposition (22 percent), and precipitation (18 percent). Ammonia is not considered an air pollutant because there are no known public health implications, and because it is a natural constituent of the atmosphere derived almost entirely from natural sources. For example, a single cow releases as much nitrogen to the atmosphere in feces and urine as 12 people would contribute if all of their ammonia production were stripped to the atmosphere.

There are no standards in the United States for ammonia concentrations in the atmosphere. Some foreign standards[1] have been established.

- Czechoslovakia, 100 mg/m^3 (24 hours)
- U.S.S.R., 200 mg/m^3 (24 hours)
- Ontario, Canada, 3,500 mg/m^3 (30 minutes)

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All of these standards are far above the 6 mg/m^3 that will occur right at the tower discharge. The process cannot be dismissed from consideration because of air pollution.

A remaining question is the fate of the ammonia discharge to the air. Is it likely to find its way into the receiving stream by being scavenged from the atmosphere by precipitation?

Ammonia may be washed from air by rainfall, but not by snowfall. The natural background concentration of ammonia in the atmosphere is 5-7 ppb. In rainfall the natural background ranges from 0.01 to 1 mg/l, with the most frequently reported values of 0.1 to 0.2 mg/l. The amount of ammonia in rainfall is related directly to the concentration of ammonia in the atmosphere. Thus, an increase in the ammonia in rainfall wuuld occur only in that area where the stripping-tower discharge increases the natural background ammonia concentration in the atmosphere.

Calculations for the ammonia washout in a rainfall rate of 3 mm/h (0.12 in./h) have been made for the Orange County, Calif., project. The ammonia concentrations of ammonia in the rainfall would approach natural background levels within 16,000 feet of the tower. Of course, the ammonia discharge during dry periods diffuses into the atmosphere quickly so that the background concentration and resulting washout rate of ammonia at greater distances from the tower are not affected during a subsequent storm. The ultimate fate of the ammonia that is washed out by rainfall within the 16,000-foot downwind distance depends on the nature of the surface upon which it falls. Most soils will retain the ammonia. That portion which lands on paved areas or directly on a stream surface will appear in the runoff from that area. Even though a portion of the ammonia washed out by precipitation will find its way into surface runoff, the net discharge of ammonia to the aquatic environment in the vicinity of the plant would be very substantially reduced.

One of the great advantages of this method of nitrogen removal is its extreme simplicity. Water is merely pumped to the top of the tower at a high pH, air is drawn through the fill, and the ammonia is stripped from the water droplets. The only control required is the proper pH in the influent water. This simplicity of operation also enhances the reliability of the process.

Several factors affect the efficiency of the ammonia-stripping process.

- Type of stripping unit
- pH
- Temperature
- Loading rate
- Scale of deposition

There are three basic types of stripping units now being used in full-scale applications.

- Countercurrent towers
- Crossflow towers
- Stripping ponds

Countercurrent towers (the entire airflow enters at the bottom of the tower while the water enters the top of the tower and falls to the bottom) have been found to be the most efficient. In the crossflow towers, the air is pulled into the tower through its sides throughout the height of the

packing. This type of tower has been found to be more prone to scaling problems. The stripping-pond approach will be discussed in more detail later.

The pH of the water has a major effect on the efficiency of the process. The pH must be raised to the point that all of the ammonium ion is converted to ammonia gas. The pH required varies somewhat with temperature,[2] but is generally about 11.0.

Another critical factor is the air temperature. The water temperature has less effect on performance because the water temperature reaches equilibrium with the air temperature in the top few inches of the stripping tower. The efficiency of the process decreases as the temperature decreases. For example, at 20° C 90 percent removal of ammonia is typically achieved. At 10° C, the maximum removal efficiency drops to about 75 percent. When air temperatures reach freezing, the tower operation must generally be shut down owing to icing problems.

The hydraulic loading rate of the tower is also an important factor. This rate typically is expressed in terms of gallons per minute applied to each square foot of the plan area of the tower packing. When the hydraulic loading rates become too high, good droplet formation is disrupted and the water begins to flow in sheets. Tower loading rates of 2 gal/min/ft^2 have been shown to be compatible with optimum tower performance.[2] It is critical that the water and air be uniformly distributed over the tower area.

Another factor that may have an adverse effect on tower efficiency is scaling of the tower packing resulting from deposition of calcium carbonate from the unstable, high-pH water flowing through the tower. The original crossflow tower at the South Lake Tahoe plant has suffered a severe scaling problem. The severity of the scaling problem was not anticipated from the pilot studies in which a countercurrent tower was used. As a result, the full-scale crossflow-tower packing was not designed with access for scale removal in mind. Thus, portions of the tower packing are inaccessible for cleaning. Those portions that were accessible were readily cleaned by high-pressure hosing. The potential scaling problem must be recognized in design. The use of countercurrent towers and design of the packing with access for cleaning can adequately combat this problem.

An example of design for scale control is the 15-mgd tower now under construction at the Orange County, Calif., Water District plant (fig. I-1). There the tower packing has been designed to be readily removable for cleaning as a precaution against scaling problems, although no significant scaling problem has been observed in several months of pilot tests at Orange County.[3] Scaling has also been reported not to be a significant problem at the Windhoek, South Africa, plant where only a soft, easily removed scale was encountered.[4] On the other hand, tests at the Blue Plains pilot plant encountered a hard scale that was extremely difficult to remove.[5] The hardness of the scale at Blue Plains was affected by operating pH, with a harder scale forming at pH 11.5 than at pH 10.8.

Typical design criteria are

- Hydraulic loading, 1 to 3 gal/min/ft^2
- Air-to-water ratio, 300 to 500 ft^3/min per gal/min
- Air-pressure drop, 0.5 to 1.25 inches water
- Fan-tip speed, 9,000 to 12,000 ft/min
- Fan-motor speed, 1 or 2 speed
- Packing depth, 20 to 25 feet

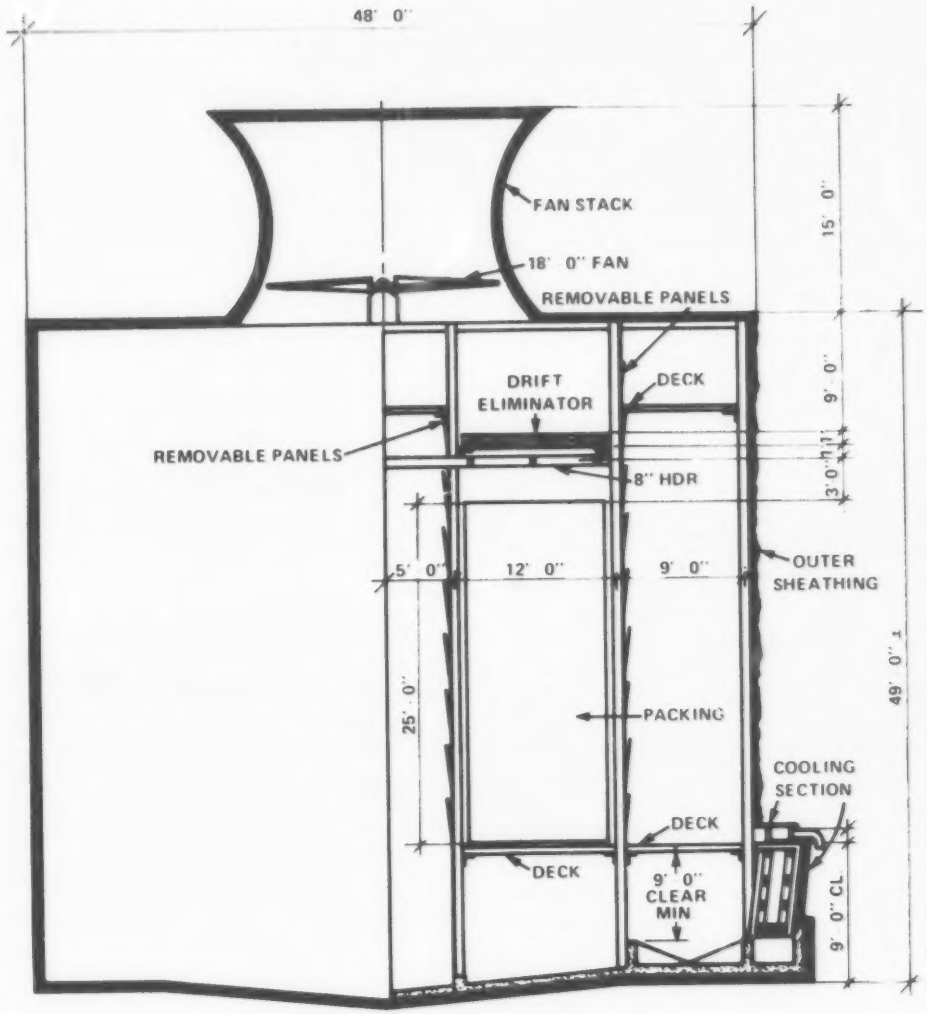

Figure I-1. Ammonia-stripping tower design, Orange County, Calif.

- Packing spacing, 2 to 4 inches horizontal and vertical
- Packing material, wood, plastic (½-in. PVC pipe being used at Orange County)

A curve for estimating the costs of the ammonia-stripping process for various-size plants is presented in figure I-2. This curve is based on a loading rate of 2 gal/min/ft^2. Because some applications may require ammonia removal only during warm weather months, operating costs are shown for both 6-month and 12-month operation.

The South Tahoe system is being modified to reduce the impact of temperature and scaling limitations encountered at this plant.[6] Basically, the modified process will consist of three steps (see figs. I-3, I-4, and I-5).

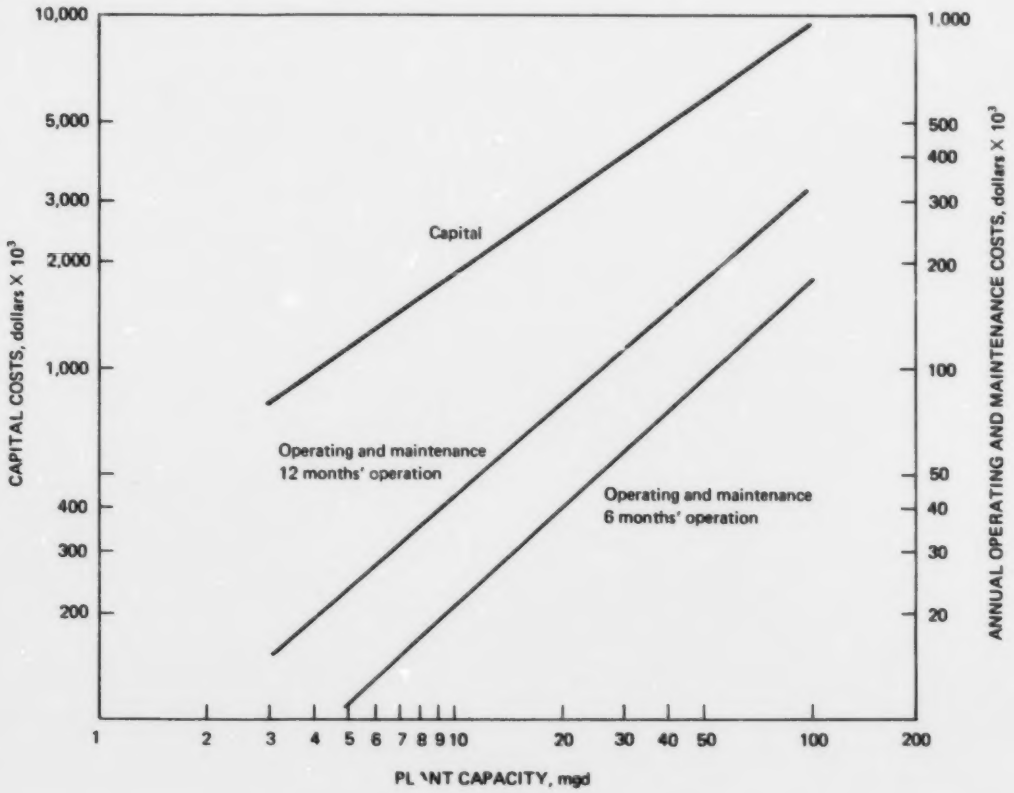

Figure I-2. Ammonia-stripping costs. (EPA STP Index = 200; includes engineering, legal, administrative, construction financing, and contingencies.)

- Holding in high-pH, surface-agitated ponds
- Stripping in a modified, crossflow forced-draft tower through air sprays installed in the tower
- Breakpoint chlorination

This system was inspired by observations in Israel of ammonia nitrogen losses from high-pH holding ponds.[7]

Pilot tests at South Tahoe indicated that the release of ammonia from high-pH ponds could be accelerated by agitation of the pond surface. In the modified Tahoe system, the high-pH effluent from the lime clarification process will flow to holding ponds. Holding pond detention times of 7-18 hours will be used in the modified South Tahoe plant. The pond contents will be agitated and recycled 4-13 times by pumping the pond contents through vertical spray nozzles into the air above the ponds. At least 37 percent ammonia removal is aniticipated, even in cold weather conditions, in the ponds. The pond contents then will be sprayed into the forced-draft tower. The packing will be removed from the tower and the entire area of the tower will be equipped with water sprays. At least 42 percent removal of the ammonia in the pond effluent is anticipated, based on pilot tests, from this added spraying in cold weather, which will include recycling of the pond effluent through the tower to achieve 2-5 spraying cycles. The ammonia escaping this process then

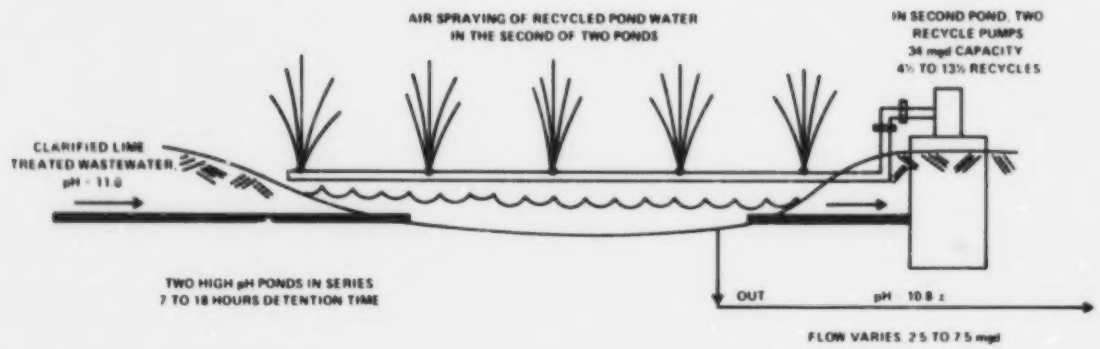

Figure I-3. Proposed new and modified ammonia nitrogen removal processes, South Lake Tahoe: New high-pH flow-equalization ponds.

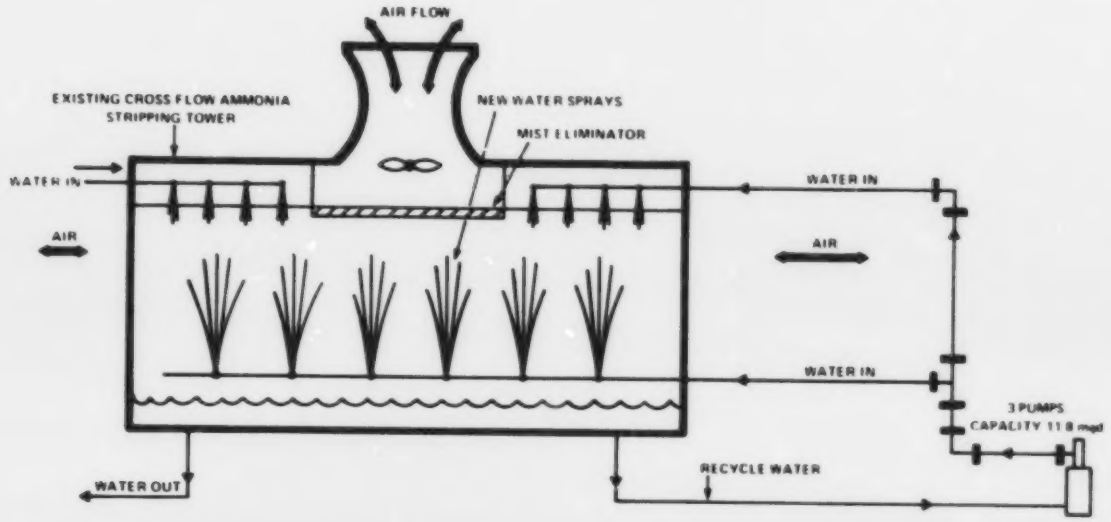

Figure I-4. Proposed new and modified ammonia nitrogen removal processes, South Lake Tahoe: Existing stripping tower modified with new sprays.

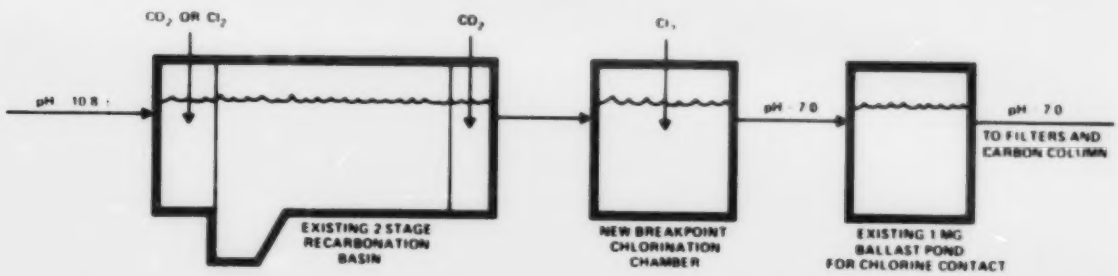

Figure I-5. Proposed new and modified ammonia nitrogen removal processes, South Lake Tahoe: Breakpoint chlorination (new).

will be removed by downstream breakpoint chlorination. The quantity of ammonia to be removed by breakpoint chlorination will vary from 5 to 16 mg/l, depending on the plant flow and temperatures.

Another approach to overcoming the limitations of the stripping process has been developed by CH2M/HILL Consulting Engineers.[8] Although the process is only in its initial stages of development, preliminary tests indicate it may be a significant advance in the state of the art of nitrogen removal. It appears that the new process overcomes most of the foregoing limitations and has the advantage of recovery of ammonia as a byproduct.

The improved process, shown diagrammatically in figure I-6, includes an ammonia-stripping unit and an ammonia-absorption unit. Both of these units are essentially sealed from the outside air but are connected by appropriate ducting. The stripping gas, which initially is air, is maintained in a closed cycle. The stripping unit operates essentially in the same manner that is now being or has been used in a number of systems, except that this system recycles the gas stream rather than using single-pass outside air.

Most of the ammonia discharged to the gas stream from the stripping unit is removed in the absorption unit. The absorbing liquid is maintained at a low pH to convert absorbed and dissolved ammonia gas to ammonium ion. This technique effectively traps the ammonia and also has the effect of maintaining the full driving force for absorbing the ammonia, since dissolved ammonia

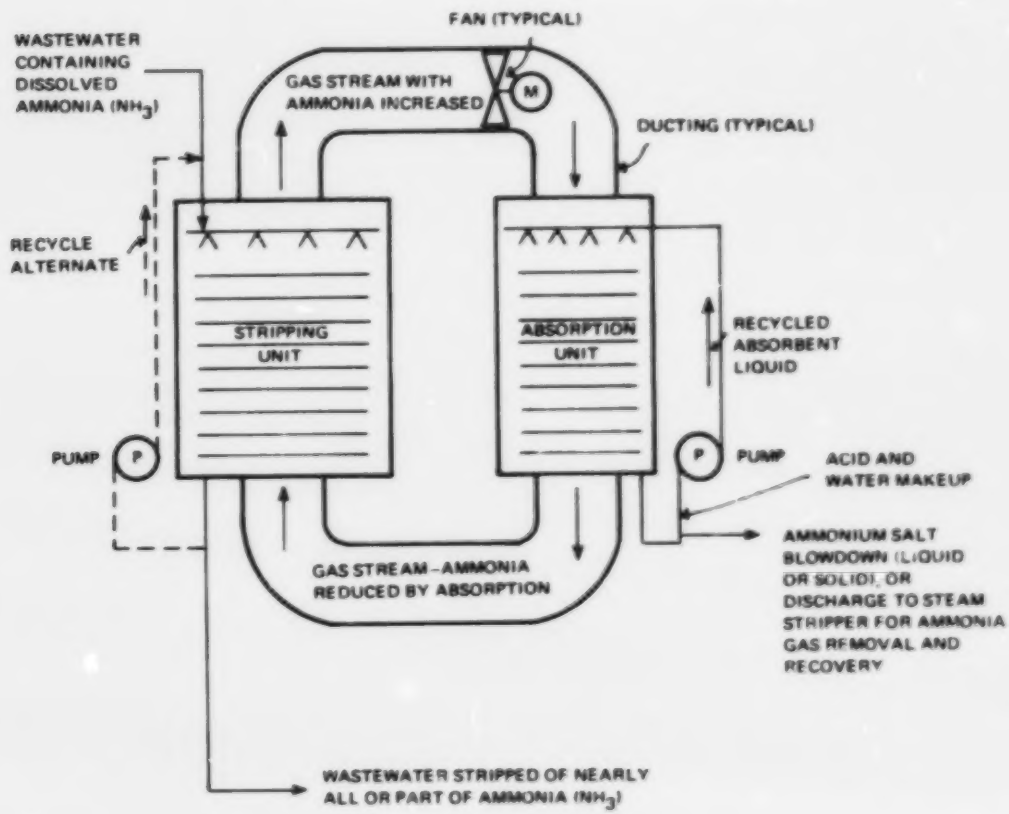

Figure I-6. Process for ammonia removal and recovery.

gas does not build up in the absorbent liquid. The absorption unit can be a slat tower, packed tower, or sprays similar to the stripping unit, but will usually be smaller owing to kinetics of the absorption process.

The absorbent liquid initially is water with acid added to obtain low pH, usually below 7.0. In the simplest case, as ammonia gas is dissolved in the absorbent and converted to ammonium ions, acid is added to maintain the desired pH. If sulfuric acid is added, for example, an ammonium sulfate salt solution is formed. This salt solution continues to build up in concentration and the ammonia is finally discharged from the absorption device as a liquid or solid (precipitate) blowdown of the absorbent. With current shortages of ammonia-based fertilizers, a salable byproduct may result.

Other methods of removal of the ammonia from the absorbent may also be applicable, depending on the acid used and the desired byproduct. Ammonia gas or aqua ammonia could be produced, for example, by steam stripping the absorbent. In this case, acid makeup would be unnecessary.

It is believed that the usual scaling problem associated with ammonia-stripping towers will be eliminated by the improved process, since the carbon dioxide which normally reacts with the calcium and hydroxide ions in the water to form the calcium carbonate scale is eliminated from the stripping air during the first few passes. The freezing problem is eliminated owing to the exclusion of nearly all outside air. The treatment system will normally operate at the temperature of the wastewater.

Chapter II

SELECTIVE ION EXCHANGE

The selective ion exchange process derives its name from the use of zeolites that are selective for ammonia relative to calcium, magnesium, and sodium. The zeolite currently favored for this use is clinoptilolite, which occurs naturally in several extensive deposits in the Western United States. Studies of the process have been conducted by Battelle Northwest[9] and the University of California.[10] Clinoptilolite used in studies conducted by Battelle Northwest for EPA was obtained from the Hector, Calif., leases of the Baroid Division of the National Lead Company, Houston, Tex. The clinoptilolite is crushed and sieved to obtain a 20 by 50 mesh size. Ammonia is removed by passing the wastewater through a bed of clinoptilolite at a rate of about 10 bed volumes per hour.

The use of clinoptilolite was investigated at the University of California with the objective of optimizing its application to ammonia removal from wastewaters. Pilot-plant operations were carried out at three different municipal sewage-treatment plants. An average ammonia removal of 96 percent was obtained in these operations with influent ammonia nitrogen concentrations of about 20 mg/l.

The ammonia capacity of the clinoptilolite was found to be nearly constant over the pH range of 4.0 to 8.0, but diminished rapidly outside this range. The effect of wastewater composition on the ammonia exchange capacity was analyzed by exhausting clinoptilolite beds with waters having different chemical compositions. For relatively constant influent ammonia concentrations, the ammonia exchange capacity was observed to decrease sharply with increasing competing action concentrations up to about 0.01 molar. Increases of cation concentrations above this value continued to decrease the exchange capacity, but to a much lesser degree. Ammonia removal to residual levels less than 0.5 mg/l ammonia nitrogen is technically feasible, but only with shorter service cycles and greater regeneration requirements. Flow rates in the range of 7.5 to 15 bed volumes per hour had no effect on ammonia effluent values.

Battelle Northwest conducted pilot studies of the clinoptilolite process applied to secondary effluents, advanced waste treatment effluents, and clarified raw sewage.[9,11] Ammonia removals ranging from 93 to 97 percent were demonstrated using a 100,000-gal/d mobile pilot plant. These studies were conducted at several different locations across the United States.

After about 150-200 bed volumes of normal-strength municipal waste have passed through the bed, the capacity of the clinoptilolite has been used to the point that ammonia begins to leak through the bed. At this point, the clinoptilolite must be regenerated so that its capacity to remove ammonia is restored.

The key to the applicability of this process is the method of handling the spent regenerant. The resin is regenerated by passing concentrated salt solutions through the exchange bed when the ammonia concentration has reached the maximum desirable level. Following regeneration, the ammonia-laden spent-regenerant volume is about 2.5 to 5 percent of the throughput treated before regeneration.

The original approach to recovering and reusing the regenerant was to use a lime slurry as the regenerant so that the ammonium stripped from the bed during regeneration would be converted to gaseous ammonia, which could then be removed from the regenerant by air stripping.[9]

Regeneration with lime alone was found to be a rather slow process; therefore, the ionic strength of the regenerant solution was increased by the addition of salt (NaCl). The increased ionic strength of the regenerant plus the presence of sodium ion accelerates the removal of ammonia from the zeolite. Although most of the sodium chloride added to the regenerant is converted to calcium chloride by continuous recycle of the regenerant, sufficient sodium ion remains under steady state conditions to promote the elution of the ammonium ions. The sodium ion has a higher diffusion coefficient than calcium ion, which is believed responsible for increasing the ammonia elution rate. With the lime-slurry regenerant, the regenerant stripping tower handles only a small fraction of the total plant throughput. Heating the stripping tower, even during cold weather periods, is then practical.

The use of the high-pH regenerant is accompanied by an operational problem. Some plugging of the bed with $Mg(OH)_2$ and $CaCO_3$ occurs when the high-pH regenerant is used. Attrition of the zeolite is aggravated by the violent backwashing needed to remove these solids, and is 0.17-0.25 percent per cycle, making makeup clinoptilolite costs a significant factor. These problems make more recently developed methods of regenerant recovery more attractive.

In one approach, ammonia in the regenerant solution may be converted to nitrogen gas by reaction with chlorine which is generated electrolytically from the chlorides already present in the regenerant solution. This process can be carried out with a regenerant of neutral pH so that the problem of precipitation of $Mg(OH)_2$ and $CaCO_3$ within the bed during regeneration is eliminated. Also, cold weather does not affect the regenerant recovery process. The regenerant solutions used are rich in NaCl and $CaCl_2$ which provide the chlorine produced at the anode of the electrolysis cell. The reactions for the destruction of ammonia by chlorine are the same as for breakpoint chlorination.

During regeneration of the ion exchange bed, a large amount of calcium is eluted from the zeolite along with the ammonia. This calcium may be removed from the spent regenerant solution by soda ash softening before passing the spent regenerant through the electrolytic cells. The softening step would lower the calcium concentration below the level that would cause calcium hydroxide formation in the electrolytic cells. High flow velocities through the electrolysis cells are required in addition to a low concentration of $MgCl_2$ to minimize scaling of the cathode by calcium hydroxide and calcium carbonate. Acid flushing of the cells would be necessary to remove this scale when the cell resistance becomes too high for economical operation.

In pilot tests of the electrolytic treatment of the regenerant at Blue Plains, Battelle Northwest found that about 50 Wh of power were required to destroy 1 gram of ammonia nitrogen (NH_3-N). When related to the treatment of water containing 25 mg/l NH_3-N, the energy consumed would be 4.7 kWh per 1,000 gallons. Tests at South Tahoe also indicated that a value of 50 Wh per gram is reasonable for design.[12] Preliminary capital and operating costs of $1.5 million and 9 cents per 1,000 gallons, respectively, were estimated by Battelle for a 10-mgd plant using electrolytic destruction of ammonia in recycled regenerant containing chloride salts of calcium, sodium, and magnesium. Electrolytic treatment of the regenerant avoids the disposal of ammonia to the atmosphere or disposal of aqueous ammonia concentrates. Total costs, including capital amortization, were estimated at 12.7 cents per 1,000 gallons.[11]

A 22.5-mgd plant designed by CH2M/HILL for the Upper Occoquan Sewage Authority in the State of Virginia will employ selective ion exchange with electrolytic treatment of the regenerant for ammonia removal. This plant will utilize soda ash softening of the regenerant to avoid cathodic scaling of the electrolysis cells. A simplified flow schematic of the regeneration system is illustrated

in figure II-1. The regeneration of the clinoptilolite beds will be accomplished with a 2-percent solution of NaCl. The spent regenerant will be collected in a large holding tank to minimize variation in the calcium content before soda ash addition for calcium removal. After the soda ash addition, the regenerant will be clarified and transferred to another holding tank where the regenerant will be recirculated through electrolysis cells for ammonia destruction.

Design criteria for the ammonia-removal plant for the Upper Occoquan District are summarized in table II-1. The electrolysis cell to be used by this plant is a 500-Ampere unit manufactured by Pacific Engineering and Production Company of Nevada, Henderson, Nev. The cell consists of a lead dioxide coated graphite anode in a cylindrical stainless steel vessel which is the cathode. The lead dioxide is highly resistant to attack by chlorine or oxychloroacids. The estimated total cost for this plant is 12.6 cents per 1,000 gallons for the selective ion exchange process.

In order to develop the design criteria for the Occoquan plant, CH2M/HILL conducted pilot tests of the process at the South Tahoe plant.[12] The ammonia concentration in the wastewater at South Tahoe ranged from 21 to 28 mg/l during these pilot tests. After about 6 weeks of pilot-plant operation, the calcium concentration of the influent increased from about 55 mg/l to about 80 mg/l. This increased calcium concentration together with concurrently occurring lower influent temperatures reduced the quantity of ammonia that could be loaded onto the clinoptilolite before a breakthrough of 1 mg/l of ammonia. The average loading to the clinoptilolite column before breakthrough of 1 mg/l of ammonia was 144 bed volumes with an influent containing 55 mg/l calcium at 22° C. When the influent calcium increased to 80 mg/l and the temperature dropped to 14° C, the loading capacity of the clinoptilolite column dropped to 104 bed volumes. Ammonia removals achieved were in excess of 95 percent.

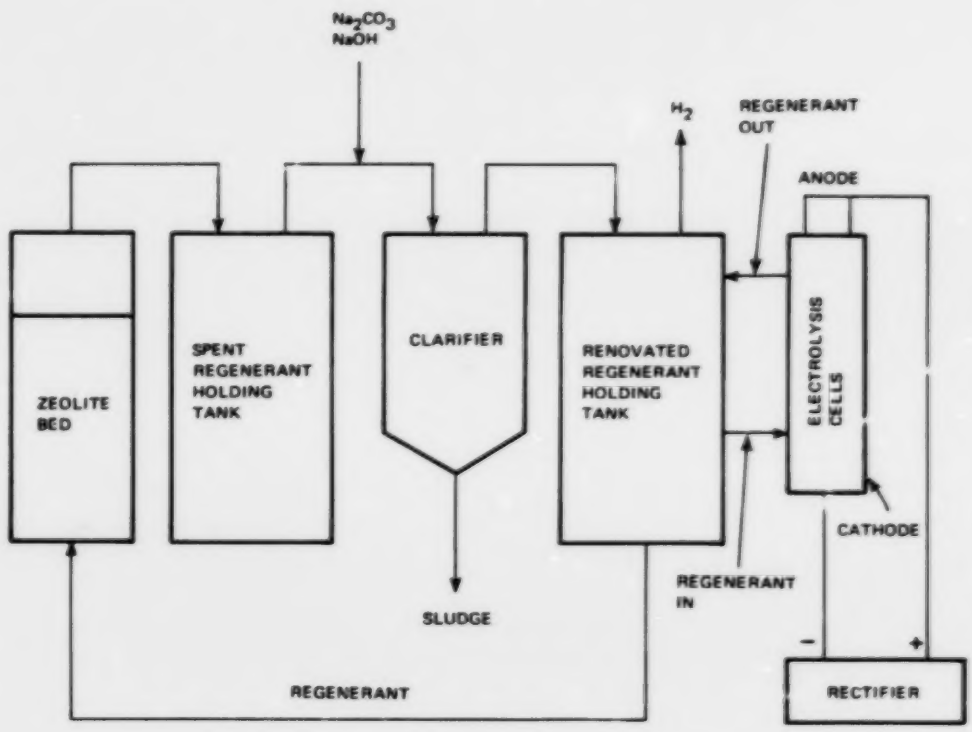

Figure II-1. Simplified flow diagram of Upper Occoquan regenerant treatment system.

Table II-1.—*Design criteria for the Upper Occoquan ammonia removal plant at 22.5-mgd flow rate*

Exchange beds:	
Size and type	10-foot-diameter X 50-foot-long horizontal pressure units
Number	8
Media	Clinoptilolite
Media size	20 X 50 mesh
Bed depth	4 feet
Bed length	50 feet
Bed width	10 feet
Service cycle loading:	
Average	9.1 BV/h
Maximum	14.1 BV/h
Hydraulic loading:	
Average	4.4 gal/min/ft^2
Maximum	6.9 gal/min/ft^2
Flow:	
Average	3.2 mgd per bed
Maximum	5 mgd per bed
Length of service cycle	200 BV
Bed loading	365 pounds NH_3 per bed cycle
Backwash water	Carbon-column effluent
Backwash rate	8 gal/min/ft^2
Exchange-bed regeneration:	
Length of cycle	3.1 hours
Regeneration rate	10 BV/h
Regenerant recovery:	
Method	Electrolysis
Power requirement	40 Wh per gram NH_3-N destroyed
NH_3 destruction rate	0.16 pound NH_3-N per hour per cell
Number of electrolytic cells in service	480
Total number of cells provided	720
Rectifiers:	
Number	3
Capacity	750 kW
Salt requirements	13,900 lb/d

The pilot column was regenerated successfully with a 2-percent sodium chloride solution at neutral pH. No loss of clinoptilolite by attrition was observed when using the neutral regenerant, and no difficulties in backwashing were observed. Although the neutral regeneration scheme was found to involve 30-40 bed volumes of regenerant rather than the 10 or less needed by others with the high-pH schemes, the minimization of attrition losses is achieved without significant disadvantage. The closed-loop regenerant-recovery system results only in added downtime for regeneration.

Scaling within the electrolytic cell used for regenerant recovery was the primary concern of the Occoquan pilot-plant study; therefore, the electrolytic cell was routinely dismantled and inspected for scaling. The flow rate through the cell was set initially at velocities of 0.13 to 0.16 ft/s, and a thin buildup of scale was observed on the cathode at the bottom-cell-inlet end after 160 hours of operation. After 230 hours of operation, the flow velocity was reduced to 0.06 ft/s, and very light scale buildup was observed depositing over the entire cathode area.

Scale was removed from a 1-in.^{2} area of the cathode, and the flow velocity through the cell was increased to 0.21 ft/s to determine the effect of scaling at higher cell velocities. At this increased flow, which was maintained for most of the period of the pilot-plant study, no new scale was deposited on the cathode. Visually, it appeared that from 25 to 50 percent of the previously deposited scale was removed. These observations suggest that scaling within the cell can be controlled by sufficient flow velocities. The average power requirements for regenerant recovery were measured as 43.3 Wh per gram ammonia destroyed. To allow for normal system losses, a design value of 50 Wh/g appears reasonable.

An alternative to air stripping or electrolysis of the regenerant is steam stripping. A 0.6-mgd plant in Rosemount, Minn., which is now entering its startup period, utilizes this technique.[13,14] At Rosemount ammonia is recovered from the spent ion exchange regenerant in an ammonia stripper. Steam is injected into a distillation column countercurrent with the regenerant solution to strip off the ammonia. An air-cooled plate-and-tube condenser then condenses the vapor for collection in a covered tank as 1-percent aqueous ammonia for sale as a fertilizer, However, it is a dilute (1 percent) ammonia solution, which reduces its potential for sale as a fertilizer, since commercial fertilizers require handling of only 1/10 the volume of liquid for the same ammonia application.

No detailed data on the Rosemount design and anticipated operating parameters were available at the time of this report. An EPA evaluation of the plant will be made in 1974 after the initial shakedown problems are resolved. The steam-stripping process is based on the use of the high-pH regenerant, which has the disadvantages noted earlier. Battelle Northwest's evaluation of steam stripping[a] indicates that it is economically feasible if the regenerant volume is held to 4 bed volumes per cycle, which is achievable with high-pH regenerant. The steam requirements were estimated to be 15 pounds per 1,000 gallons. At a steam cost of $2 per 1,000 pounds, the steam costs would be only 0.03 cent per 1,000 gallons. Heat recovery by contacting the cold regenerant with stripped regenerant and by contacting it with the condenser would be necessary to achieve economical operation. Because of the unstable, high-pH regenerant, scaling problems on the heat exchanges could be anticipated.

Another technique for regenerant recovery is the use of the stripping-recovery process (shown in fig. I-4) on the spent regenerant. A 6-mgd plant at North Lake Tahoe is being designed using this approach. Tests to date indicate that ammonia sulfate concentrations of 50 percent are readily achievable in the absorption tower. The estimated costs of the selective ion exchange approach based on this technique of regenerant recovery are shown in figure II-2. No credit for potential sale of ammonium sulfate has been included.

[a]B. W. Mercer, Battelle Northwest, personal communication, Dec. 14, 1973.

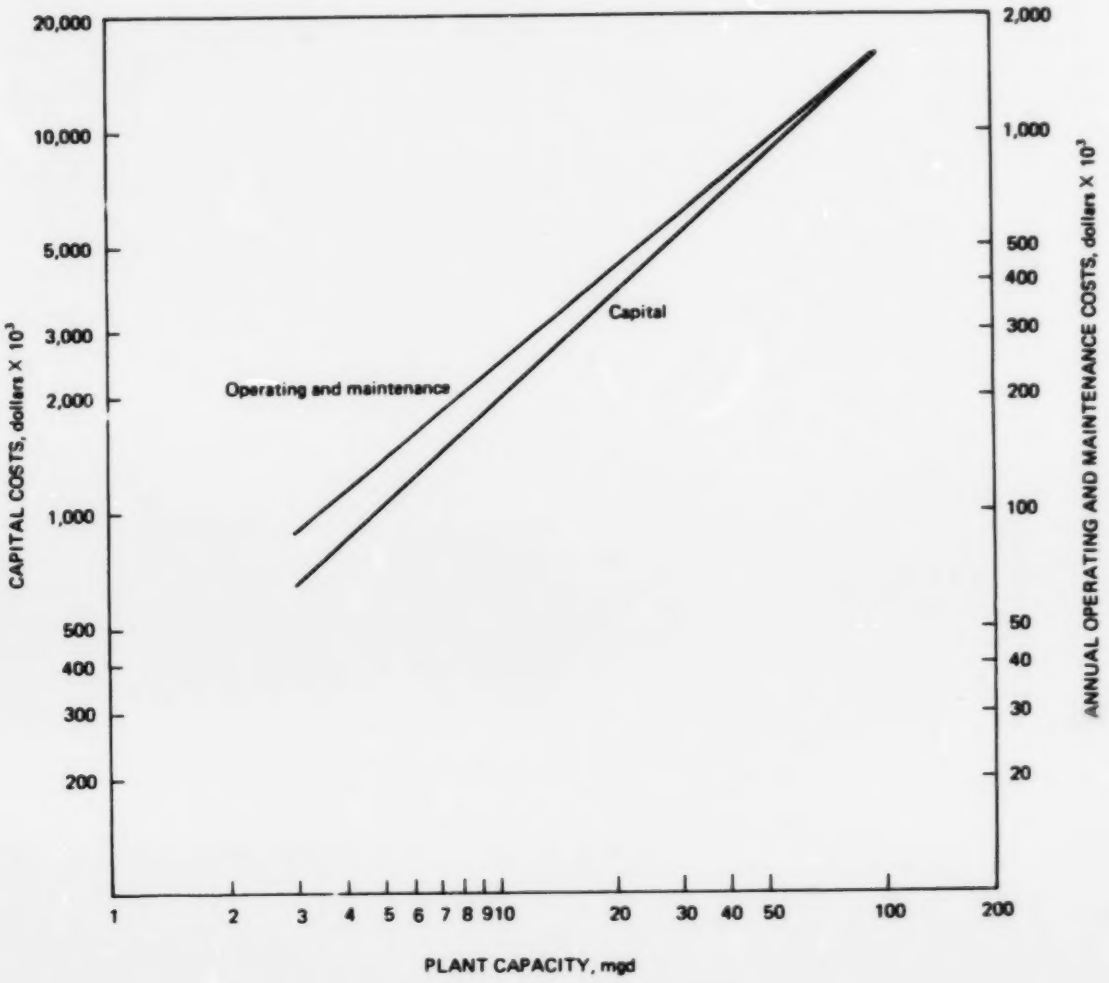

Figure II-2. Ammonia removal by selective ion exchange. (EPA STP Index = 200; includes engineering, legal, administrative, construction financing, and contingencies.)

Chapter III

BREAKPOINT CHLORINATION

When chlorine is added to a wastewater containing ammonia nitrogen, ammonia reacts with the hypochlorous acid formed to produce chloramines. Further addition of chlorine to the breakpoint converts the chloramines to nitrogen gas. The chlorine and ammonia reactions in dilute solutions are

$$NH_4 + HOCl \rightarrow NH_2Cl \text{ (monochloramine)} + H_2O + H^+$$

$$NH_2Cl + HOCl \rightarrow NHCl_2 \text{ (diochloramine)} + H_2O$$

$$NCHl_2 + HOCl \rightarrow NCl_3 \text{ (nitrogen trichloride)} + H_2O$$

The reactions are dependent on pH, temperature, contact time, and initial chlorine-to-ammonia ratio. Chlorine is added to the wastewater being treated until the chlorine residual has reached a minimum (the breakpoint) and the ammonia is removed. A typical breakpoint curve is shown in figure III-1. The reaction with ammonia is very rapid. Less than 1 minute, in the pH range of 7.0 to 8.0, and all of the free chlorine is converted to monochloramine at a 5:1 weight ratio of chlorine:ammonia nitrogen. As the weight ratio exceeds 5:1, the monochloramine breaks down and forms dichloramine and ammonia,

$$2NH_2Cl \rightarrow NHCl_2 + NH_3$$

Monochloramine is then oxidized by excess chlorine under slightly alkaline conditions to nitrogen gas,

$$2NH_2Cl + HOCl \rightarrow N_2\uparrow + 3HCl + H_2O$$

Stoichiometrically, a weight ratio of 7.6:1 of chlorine to ammonia nitrogen is required to oxidize ammonia to nitrogen gas.

Breakpoint chlorination tests on domestic wastewaters at the Blue Plains plant indicate that 95 to 99 percent of the ammonia is converted to nitrogen gas and that no significant amount of nitrous oxide is formed.[15] The quantity of chlorine required to achieve breakpoint was found to decrease with an increasing degree of treatment before the breakpoint process. The quantity of chlorine required for breakpoint chlorination of raw wastewater was found to be 10 parts by weight of Cl_2 to 1 part of NH_3 nitrogen. This ratio decreased to 9:1 Cl_2:NH_3 nitrogen for secondary effluents, and 8:1 Cl_2:NH_3 nitrogen for lime-clarified and filtered secondary effluent. The Blue Plains tests found that the chlorine dose was minimized at pH values between 6.0 and 7.0. The minimum NO_3 production (1.5 percent of the NH_3-N) occurred at pH 5.0. At pH 8.0, the nitrate production increased to 10 percent of the influent NH_3 nitrogen. NCl_3 production at the breakpoint decreased from 1.5 percent to the influent at pH 5.0 to 0.25 percent at pH 8.0. Temperature did not affect the product distribution or the required chlorine dose in the range 5° to 40° C.

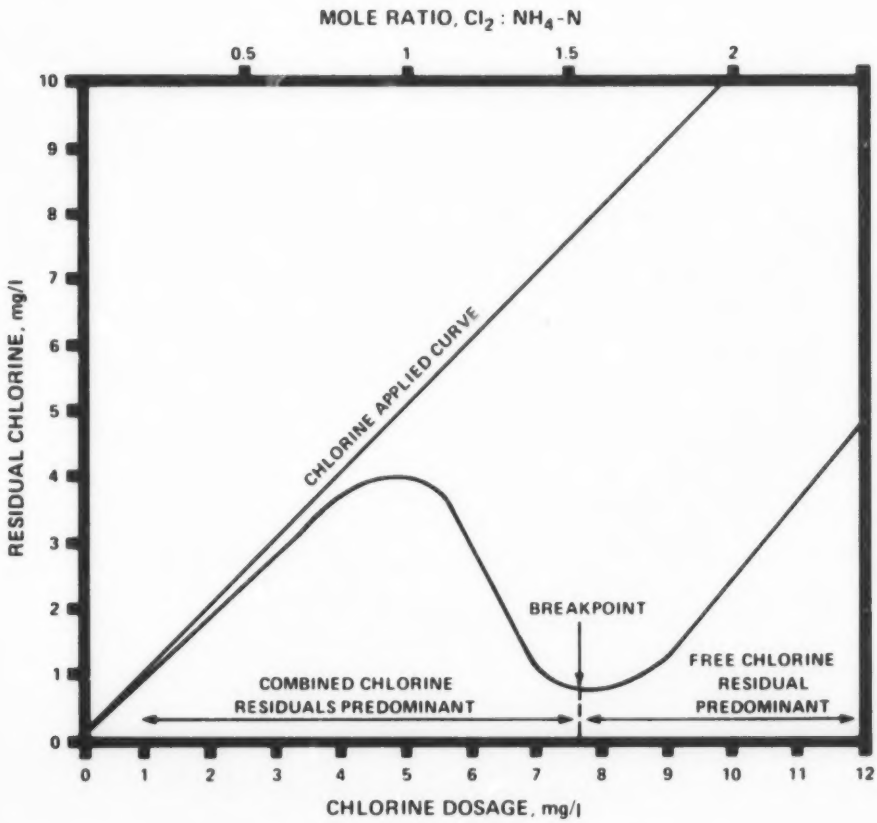

Figure III-1. Typical breakpoint-chlorination curve.

The use of chlorine produces an equivalent weight of hydrochloric acid which may depress the pH of the wastewater unless the natural alkalinity is adequate or a base such as sodium hydroxide is added. If the pH is allowed to fall, highly odorous nitrogen trichloride (NCl_3) is formed, which is an intolerable end product. If a base is used to prevent pH depression, the mixing of the wastewater, chlorine, and base must be extremely violent to avoid local areas of low pH which would generate NCl_3. Tests at Blue Plains showed that eductors do not give adequate chlorine-wastewater mixing, which did result in localized low-pH regions in which objectionable quantities of NCl_3 formed. Violent mechanical mixing is required. The use of sodium hypochlorite rather than chlorine does not depress the pH and avoids the foregoing problem.

The use of chlorine gas may produce more acid than can be neutralized by the wastewater. According to the EPA study reported by Pressley,[15] 14.3 mg/l of alkalinity (as $CaCO_3$) are required to neutralize the acid produced by the oxidation of 1 mg/l NH_3-N to N_2. Either sodium hydroxide or lime may be used for pH control if the wastewater is deficient in alkalinity. A wastewater containing 25 mg/l NH_3-N requires an alkalinity of about 357 mg/l if chlorine gas is used.

A significant factor in considering this process for application in some cases is the addition of dissolved solids inherent to the process. If, for example, chlorine gas were used and the influent ammonia nitrogen concentration were 25 mg/l, the dissolved solids would be increased by 156 mg/l. Neutralizing with lime would result in a total increase of 306 mg/l of total solids. If the chlorinating agent were sodium hypochlorite, the increase in dissolved solids would be 177 mg/l.[16]

The effects of breakpoint chlorination on organic nitrogen are somewhat uncertain. The Blue Plains tests[15] found only a "slight reduction in organic nitrogen within the two hour contact time." Other tests[17] observed a decrease in organic nitrogen content as the Cl_2:N ratio increased. Reductions from 3.2-3.5 mg/l to 0.2-0.4 mg/l organic nitrogen were reported for the breakpoint process. The authors,[17] however, felt that such apparent removals result from an analytical anomaly in which the organochloramine formed is not measured as nitrogen in the Kjeldahl organic nitrogen analysis. At higher chlorine dosages, however, their literature review indicated that organochloramines will be oxidized to aldehydes and nitrogen gas. The breakpoint reactions of organochloramines proceed more slowly than the ammonia chloramines, and probably will not be complete in a 30-minute contact time.

Several recent studies[16,17,18,19] have investigated the possibility of adding only enough chlorine to form monochloramines and then removing the monochloramines on activated carbon. Some advantages would be realized if monochloramine could be removed by activated carbon. The theoretical Cl:N ratio for 100 percent ammonia removal would drop from 7.6:1 for breakpoint to about 5:1 for the formation of monochloramine. The dissolved solids added to the system and the alkalinity requirements would be significantly reduced. Two studies[16,17] found that ammonia removals of about 50 percent could be achieved at Cl:N ratio of 5:1 when the breakpoint process was followed by activated-carbon adsorption. Complete removal still required dosages of about 9:1 in three studies.[16,17,18] Carbon contact times of 10 minutes were found to be adequate for complete dechlorination of the effluent.[16]

Experiences with the breakpoint process in South Africa[20] confirm that automatic control of the process is important. The African researchers concluded that monitoring of the ammonia coupled with automatically controlled chlorine dosing is a necessity. A successful, automated-computer-control system has been developed and demonstrated at the Blue Plains pilot plant.[21] This system matches the quantity of chlorine fed to the quantity of incoming nitrogen, and also controls the pH to 7.0 to minimize the formation of NCl_3 and NO_3. (See fig. III-2.)

There are several projects in the design or construction stage utilizing the breakpoint-chlorination process. The 7.5-mgd South Lake Tahoe plant is adding facilities to provide breakpoint chlorination of the quantities of ammonia which escape the upstream nitrogen-removal processes (5-16 mg/l).[6] The Orange County, Calif., 15-mgd wastewater reclamation plant now nearing completion will include facilities to remove the 2-3 mg/l of ammonia that will escape the upstream ammonia-stripping process.[22] Chlorine gas will be supplied from purchased 1-ton cylinders and by an on-site electrolytic generator rated at 2,000 lb/d. The chlorine generation system will utilize an electrochemical cell to electrolyze sodium chloride brine to chlorine gas and sodium hydroxide solution. The sodium hydroxide solution will be used in an adjacent sea water desalting plant.

A 60-mgd facility is under design for Montgomery County, Md., by CH2M/HILL, which will utilize the breakpoint process as the primary nitrogen-removal process. In this plant, sodium hypochlorite will be produced on site by electrolysis of a salt brine. The Cortland, N.Y., 10-mgd physical-chemical plant design includes facilities for breakpoint chlorination of the portion of the flow required to meet stream standards.

The costs of the process applied to the 309-mgd plant at Blue Plains were estimated at 6.7 cents per 1,000 gallons, with chemical costs constituting 5.9 cents of this value. These costs were based on a chlorine cost of only $75 per ton and a dose of only 120 mg/l. The control of pH was assumed to be by lime addition (1 pound of lime per pound of chlorine) at a lime cost of $24 per ton. In any case, the cost of the chlorine itself constitutes a large portion of the total project costs. Assuming a chlorine cost of 0.07 cent per pound and a Cl:N ratio of 8:1, the chlorine cost for removal of 25 mg/l ammonia would be 11.8 cents per 1,000 gallons. The chlorine demand for this dose is equivalent to 1,668 lb/mg.

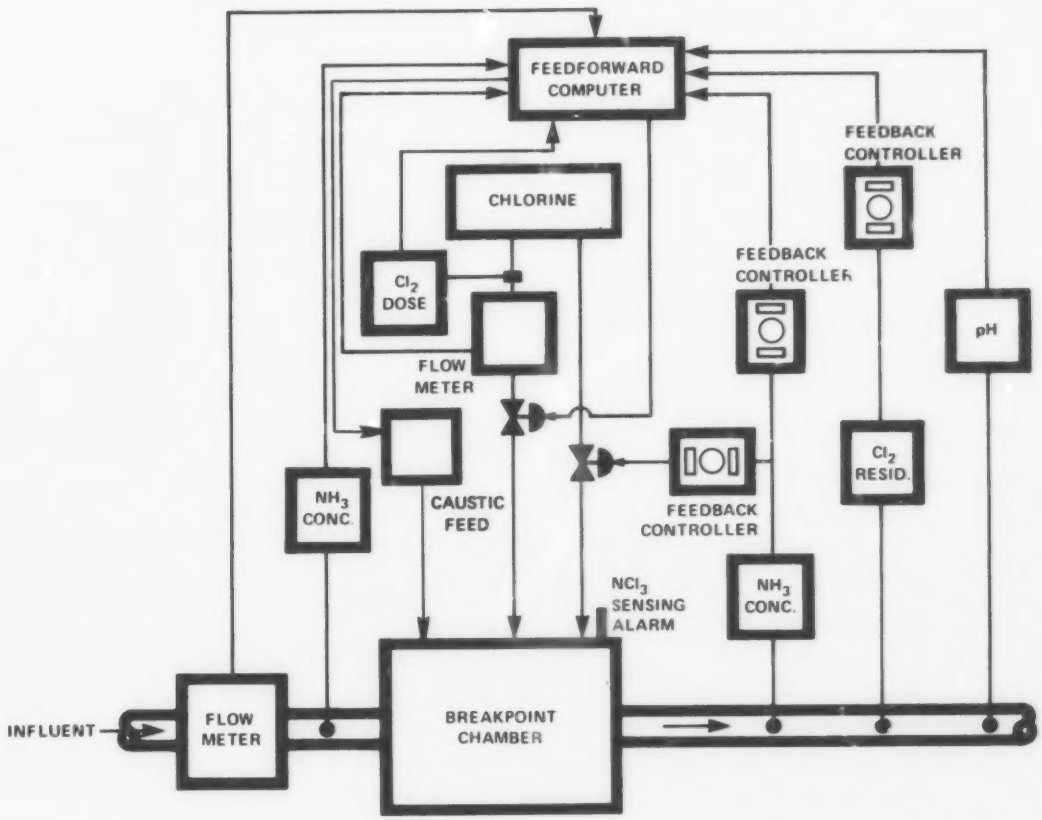

Figure III-2. Breakpoint-chlorination control system.

The breakpoint process is useful for eliminating low concentrations of ammonia as a polishing step following another nitrogen-removal process.

Chapter IV
COMPARISON OF PROCESSES

Each of the processes discussed earlier has its advantages and disadvantages. Unfortunately, no single process for nitrogen removal is superior to others both in terms of performance and economics.

The ammonia-stripping process has the advantages of low cost, removal of ammonia with a minimal addition of dissolved solids, simplicity, and reliability. However, it has the disadvantages of poor efficiency in cold weather and the potential for scaling problems that may reduce its efficiency, and it raises concerns, whether valid or not, over ammonia gas discharge. The new stripping-recovery system overcomes many of these problems, but at the sacrifice of low process costs.

The selective ion exchange process has the advantages of high efficiency, insensitivity to temperature fluctuations, removal of ammonia with a minimal addition of dissolved solids, and the ability to eliminate any discharges of nitrogen to the atmosphere other than nitrogen gas. This process has the disadvantage of relatively high cost, and process control and operation are relatively complex.

The breakpoint chlorination process has the advantages of low capital cost, a high degree of efficiency and reliability, insensitivity to cold weather, and the release of nitrogen as nitrogen gas. It has the disadvantage of adding a substantial quantity of dissolved solids to the effluent in the process of removing the ammonia, it will raise public concerns over handling of chlorine gas, the process controls required are relatively complex, and it requires a downstream dechlorination process.

The relative costs of the physical-chemical nitrogen processes for a 10-mgd plant are

- Ammonia stripping, 5 cents per 1,000 gallons
- Selective ion exchange, 10-13 cents per 1,000 gallons
- Breakpoint chlorination, 11 cents per 1,000 gallons

These costs all are based on the removal of 25 mg/l ammonia nitrogen. The cost of biological nitrogen removal by the three-stage activated-sludge process has been estimated[23,24] at about 13 cents per 1,000 gallons. Preliminary estimates on the costs of the new ammonia-stripping/ammonia-recovery process discussed earlier, which minimizes the seasonal restrictions on the ammonia-stripping process, indicate that the cost will be 8-10 cents per 1,000 gallons. It can be seen from the above costs that there is little economic incentive to select one process over another if faced with a requirement for cold weather removal of ammonia. The choice must be made by weighing the advantages and disadvantages of each approach in light of the circumstances applicable to a specific project.

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METRIC CONVERSION TABLES

Recommended Units

Description	Unit	Symbol	Comments	Customary Equivalents
Length	metre	m	*Basic SI unit*	39.37 in.=3.28 ft=1.09 yd
	kilometre	km		0.62 mi
	millimetre	mm		0.03937 in.
	micrometre	μm		$3.937 \times 10^{-2}=10^{3}$A
Area	square metre	m^2		10.764 sq ft = 1.196 sq yd
	square kilometre	km^2		0.386 sq mi = 247 acres
	square millimetre	mm^2		0.00155 sq in.
	hectare	ha	The hectare (10 000 m^2) is a recognized multiple unit and will remain in international use.	2.471 acres
Volume	cubic metre	m^3		35.314 cu ft = 1.3079 cu yd
	litre	l	The litre is now recognized as the special name for the cubic decimetre.	1.057 qt = 0.264 gal = 0.81×10^{-6} acre-ft
Mass	kilogram	kg	*Basic SI unit*	2.205 lb
	gram	g		0.035 oz = 15.43 gr
	milligram	mg		0.01543 gr
	tonne or	t	1 tonne = 1 000 kg	0.984 ton (long) =
	megagram	Mg	1 Mg = 1 000 kg	1.1023 ton (short)
Time	second	s	*Basic SI unit*	
	day	d	Neither the day nor the year is an SI unit but both are important.	
	year	year		
Force	newton	N	The newton is that force that produces an acceleration of 1 m/s^2 in a mass of 1 kg.	0.22481 lb (weight) = 7.233 poundals
Moment or torque	newton metre	N-m	The metre is measured perpendicular to the line of action of the force N. Not a joule.	0.7375 ft-lbf
Stress	pascal	Pa		0.02089 lbf/sq ft
	kilopascal	kPa		0.14485 lbf/sq in

Recommended Units

Description	Unit	Symbol	Comments	Customary Equivalents
Velocity linear	metre per second	m/s		3.28 fps
	millimetre per second	mm/s		0.00328 fps
	kilometres per second	km/s		2.230 mph
angular	radians per second	rad/s		
Flow (volumetric)	cubic metre per second	m^3/s	Commonly called the cumec	15,850 gpm = 2.120 cfm
	litre per second	l/s		15.85 gpm
Viscosity	pascal second	Pa-s		0.00672 poundals/sq ft
Pressure	newton per square metre or pascal	N/m^2 Pa		0.000145 lb/sq in
	kilometre per square metre or kilopascal	kN/m^2 kPa		0.145 lb/sq in.
	bar	bar		14.5 lb/sq in.
Temperature	Kelvin	K	*Basic SI unit*	
	degree Celsius	C	The Kelvin and Celsius degrees are identical. The use of the Celsius scale is recommended as it is the former centigrade scale.	$\frac{5F}{9} - 17.77$
Work, energy, quantity of heat	joule	J	1 joule = 1 N-m where metres are measured along the line of action of force N.	2.778×10^{-7} kw-hr = 3.725×10^{-7} hp-hr = 0.73756 ft-lb = 9.48×10^{-4} Btu
	kilojoule	kJ		2.778 kw-hr
Power	watt	W	1 watt = 1 J/s	
	kilowatt	kW		
	joule per second	J/s		

Application of Units

Description	Unit	Symbol	Comments	Customary Equivalents
Precipitation, run-off, evaporation	millimetre	mm	For meteorological purposes it may be convenient to measure precipitation in terms of mass/unit area (kg/m^2). 1 mm of rain = 1 kg/m^2	
River flow	cubic metre per second	m^3/s	Commonly called the cumec	35.314 cfs
Flow in pipes, conduits, channels, over weirs, pumping	cubic metre per second	m^3/s		
	litre per second	l/s		15.85 gpm
Discharges or abstractions, yields	cubic metre per day	m^3/d	1 l/s = 86.4 m^3/d	1.83×10^{-3} gpm
	cubic metre per year	m^3/year		
Usage of water	litre per person per day	l/person day		0.264 gpcd
Density	kilogram per cubic metre	kg/m^3	The density of water under standard conditions is 1 000 kg/m^3 or 1 000 g/l or 1 g/ml.	0.0624 lb/cu ft

Application of Units

Description	Unit	Symbol	Comments	Customary Equivalents
Concentration	milligram per litre	mg/l		1 ppm
BOD loading	kilogram per cubic metre per day	kg/m^3d		0.0624 lb/cu-ft day
Hydraulic load per unit area; e.g. filtration rates	cubic metre per square metre per day	m^3/m^2d	If this is converted to a velocity, it should be expressed in mm/s (1 mm/s = 86.4 m^3/m^2 day).	3.28 cu ft/sq ft
Hydraulic load per unit volume; e.g., biological filters, lagoons	cubic metre per cubic metre per day	m^3/m^3d		
Air supply	cubic metre or litre of free air per second	m^3/s l/s		
Pipes				
diameter	millimetre	mm		0.03937 in.
length	metre	m		39.37 in. = 3.28 ft
Optical units	lumen per square metre	lumen/m^2		0.092 ft candle/sq ft